## REMARKS

Claims 1-3, 6-15 and 17-26 are pending in this application. Claims 4, 5 and 16 have been canceled herein without prejudice or disclaimer. Claims 1, 6, 11, 14, 15, 17 and 26 have been amended herein. Applicants submit that no new matter has been added by this amendment. Minor amendments have been made in the specification.

The amendment to claim 1 incorporates the limitations of canceled claims 4 and 16. Claims 6, 11 and 14 are accordingly amended to depend from claim 1.

The amendment to claim 15 incorporates the limitations of canceled claims 4 and 16. A minor amendment is also made to correct the multiple dependency wording. Claim 17 is accordingly amended to depend from claim 15.

The amendment to claim 26 corrects the multiple dependency wording.

An amendment to Table 1 of the specification is made. Referring for convenience to the publication of the present application, U.S. Patent Publication No. 2003/0003340, as the specification of the present application, it can be seen in paragraphs [0222] to [0224] that component (B) for Comparative Example 1 was "polytetramethylene oxide".

An amendment is made to the specification on page 31, line 6, where the symbol " ≤ ", that is, "less than or equal to", has been replaced by the symbol " > ", that is, "greater than". Applicants submit that it is clear from the context of the paragraph, in which the situation of "less than or equal to" is being contrasted with the situation of "greater than", that this represents correction of a typographical error.

**General remarks.**

As a result of this amendment, the proton-containing membrane of the present invention is a proton-conducting membrane, comprising a three-dimensionally crosslinked silicon-oxygen structure (A), carbon-containing compound (B) bound to (A) via a covalent bond, and inorganic acid (C), characterized by

a phase-separated structure containing a carbon-containing phase containing at least 80% by volume of the carbon-containing compound (B) and inorganic phase containing at least 80% by volume of the inorganic acid (C), the inorganic phase forming the continuous ion-conducting paths, wherein said carbon-containing compound (B) is characterized by the skeleton section substituted with hydrogen at the joint with the three-dimensionally crosslinked silicon-oxygen structure (A), satisfying the following relationship:

$(\delta p^2 + \delta h^2)^{1/2} \leq 7\ (\mathrm{Mpa})^{1/2}$

wherein $\delta p$ and $\delta h$ are the polarity and hydrogen bond components for the three-component solubility parameter.

Further, as a result of the above-mentioned amendment, the method for producing the proton-conducting membrane of the present invention is a method for producing the proton-conducting membrane of one of Claims 1 to 3 comprising a three-dimensionally crosslinked silicon-oxygen structure (A), carbon-containing compound (B) bound to (A) via a covalent bond, and inorganic acid (C),

said method comprising steps of preparing a mixture of a carbon-containing compound (D) having one or more hydrolyzable silyl groups and said inorganic acid (C), forming the above mixture into a film, and hydrolyzing/condensing the hydrolyzable silyl group contained in the mixture formed into the film, to form said three-dimensionally crosslinked silicon-oxygen structure (A),

wherein the skeleton section of said carbon-containing compound having one or more hydrolyzable silyl groups (D) whose hydrolyzable silyl group(s) are substituted by hydrogen satisfies the following relationship:

$(\delta p^2 + \delta h^2)^{1/2} \leq 7(\mathrm{Mpa})^{1/2}$

wherein, $\delta p$ and $\delta h$ are the polarity and hydrogen bond components of the three-component solubility parameter.

As is apparent from the description in paragraph [0036] of U.S. Patent Application Publication No. 2003/0003340 A1 (referring, to convenience, to the published present application as the "specification"), the proton-conducting membrane of the present invention exhibits various excellent properties (especially, excellent proton conductivity at high temperature), since it satisfies the above the above-mentioned requirements.

In the proton-conducting membrane of the present invention comprising a three-dimensionally crosslinked silicon-oxygen structure (A), carbon-containing compound (B) bound to (A) via a covalent bond, and inorganic acid (C) (hereinbelow simply referred to as components (A), (B) and (C), respectively), the covalent bond between components (A) and (B) is extremely important for imparting high heat resistance to the proton-conducting membrane (see paragraph

[0097] of the specification).

In particular, in many cases, the organic compound preferred as component (B) is a paraffin compound or the like which is gaseous, liquid or molten at high temperature, and such a compound is inapplicable to a proton-conducting membrane serviceable at high temperature unless the above-mentioned covalent bond is formed (see paragraph [0118] and [0119] of the specification).

Further, in the proton-conducting membrane of the present invention, it is also extremely important that in the skeleton section of component (B) (i.e., component (B) substituted with hydrogen at the joint with component (A)), the relationship $(\delta p^2 + \delta h^2)^{1/2} \leq 7(\mathrm{Mpa})^{1/2}$ (wherein $\delta p$ and $\delta h$ are the polarity and hydrogen bond components of the three-component solubility parameter, respectively) is satisfied.

In the proton-conducting membrane of the present invention, an inorganic acid (i.e., component (C)) is used as the component responsible for proton conduction (agent to impart proton conductivity), and a carbon-containing compound (i.e., component (B)) is used as the component responsible for securing the membrane properties (see paragraph [0077] of the specification). In order to increase the proton conductivity of the proton-conducting membrane without deteriorating the membrane properties, it is necessary to form a phase-separated structure in which components (B) and (C) cause phase separation and component (C) forms a continuous phase. In order to achieve the formation of such a phase-separated structure, the solubility of component (B) and component (C) in each other (in other words, the compatibility between components (B) and (C)) is important (see paragraphs [0104] and [0105] of the specification).

The inventors have intensively and extensively studied the relationship between $(\delta p^2 + \delta h^2)$ value for the skeleton section of component (B) and the formation of the phase-separated structure. As a result, it has been found that when $(\delta p^2 + \delta h^2)^{1/2} > 7(\text{Mpa})^{1/2}$, dissolution (compatibilization) of components (B) and (C) starts and this makes it difficult to form the phase-separated structure, whereas when $(\delta p^2 + \delta h^2)^{1/2} \leq 7(\text{Mpa})^{1/2}$, the phase-separated structure is formed (particularly notably when $(\delta p^2 + \delta h^2)^{1/2} \leq 5(\text{Mpa})^{1/2}$) to form the continuous structure of component (C), thus improving conductivity (see paragraph [0111] of the specification). That is, the inventors have found, for the first time, an unexpected effect that the reduction of the compatibility of component (B) with component (C) increases the proton-conductivity of the proton-conducting membrane.

In fact, when the $(\delta p^2 + \delta h^2)^{1/2}$ value for the skeleton section of component (B) is larger than 7, the proton conductivity of the proton-conducting membrane is lowered due to the difficulty in forming the phase-separated structure by components (B) and (C), whereas when the $(\delta p^2 + \delta h^2)^{1/2}$ value for the for the skeleton section of component (B) is 7 or less, components (B) and (C) easily form the phase-separated structure and the proton conductivity is increased. This is apparent from the comparison between Example 1 and Comparative Example 1 (the results thereof are shown in Table 1).

In Example 1, octane ($(\delta p^2 + \delta h^2)^{1/2} = 0$) and tungstophosphoric acid are used as components (B) and (C), respectively, and the proton conductivity of the resultant membrane is $3.4 \times 10^{-2}$ s/cm at 60°C and $1.5 \times 10^{-2}$ s/cm at 140°C (see Table 1). Further, the analysis of the resultant membrane by means of a field emission type electron microscope showed the formation of the phase-separated

structure in which components (B) and (C) cause phase separation and component (C) forms a continuous phase (more specifically, the sea-island structure with the inorganic acid phase as the continuous phase) (see paragraph [0220] of the specification).

On the other hand, in the proton-conducting membrane of Comparative Example 1, polytetramethylene glycol ($(\delta p^2 + \delta h^2)^{1/2} > 7$) and tungstophosphoric acid are used as components (B) and (C), respectively, and the proton conductivity of the resultant membrane is $1.1 \times 10^{-4}$ s/cm at 60°C and $1.5 \times 10^{-3}$ s/cm at 140°C (see Table 1). Further, the analysis of the resultant membrane by means of a field emission type electron microscope showed that components (B) and (C) are almost uniformly dispersed (see paragraph [0228] of the specification).

As is apparent from the above, the two requirements that:

components (A) and (B) are bound via a covalent bond; and

$(\delta p^2 + \delta h^2)^{1/2}$ value for the skeleton section of component (B) is 7 or less,

especially the latter requirement, are extremely important in the present invention.

**VI. Claims 1-26 are rejected under the judicially created doctrine of obviousness-type double patenting as being unpatentable over claims 1-20 of U.S. Patent No. 6,680,138 (Honma et al.).**

Claim 1 of U.S. Patent No. 6,680,138 (Honma et al.) is directed to a proton-conducting membrane, comprising

(A) an organic material having a main chain, which comprises a polymethylene oxide and a

polymethylene;

(B) a three-dimensionally crosslinked structure containing a specific metal-oxygen bond;

(C) an agent for imparting proton conductivity; and

(D) water, wherein

(i) said organic material (A) has a number-average molecular weight of 56 to 30,000, and at least 4 carbon atoms connected in series in the main chain; and

(ii) said organic material (A) and three-dimensionally crosslinked structure (B) are bound to each other via a covalent bond.

It is apparent that organic material (A), three-dimensionally crosslinked structure (B) and proton conductivity imparting agent (C) in Honma et al. correspond to components (B), (A) and (C) in the present invention, respectively (although not specified by the Examiner in the Office Action).

It is conceded that Honma et al. disclosed the covalent bond between organic material (A) and three-dimensionally crosslinked structure (B). However, in Honma et al., there is no description with respect to the compatibility between organic material (A) and proton conductivity imparting agent (C) (more specifically, the requirement that $(\delta p^2 + \delta h^2)^{1/2}$ value for the skeleton section of organic material (A) is 7 or less). That is, in Honma et al., the importance of this compatibility is not recognized at all.

As apparent from the above, Honma et al. does not teach or suggest the essential features and the unexpected effects of the present invention.

**VII. Claims 1-26 with main claim 1 as the main invention are rejected under 35 U.S.C. 103(a) as being unpatentable over Takada et al. (U.S. Patent No. 5,682,261).**

Claim 1 of U.S. Patent No. 5,682,261 (Takada et al.) is directed to a protonic conductor comprising silicon oxide doped with a Brønsted acid and a thermoplastic elastomer.

It is apparent that the silicon oxide, thermoplastic elastomer and Brønsted acid in Takada et al. correspond to components (A), (B) (exactly the skeleton section of component (B)) and (C) in the present invention, respectively (although not specified by the Examiner in the Office Action).

However, in Takada et al., there is no description with respect to the covalent bond between the silicon oxide and thermoplastic elastomer.

Further, in Takada et al., there is no description with respect to the compatibility between the thermoplastic elastomer and Brønsted acid (more specifically, the requirement that $(\delta p^2 + \delta h^2)^{1/2}$ value for the thermoplastic elastomer is 7 or less). That is, in Takada et al., the importance of this compatibility is not recognized at all.

As is apparent from the above, Takada et al. does not teach or suggest that essential features and the unexpected effects of the present invention.

**VIII. Claims 1-26 with main claim 1 as the main invention are rejected under 35 U.S.C. 103(a) as being unpatentable over Fenton et al. (U.S. Patent No. 6,465,136). Kim et al. US 2003/0099874 is cited to show the state of the art.**

Applicants respectfully note that Kim et al. '874 is not prior art against the present

application, since it was filed on August 15, 2002, after the filing date of the present application. As such, it is unclear to Applicants how Kim et al. "shows the state of the art" at the time of filing of the present application.

Claim 1 of U.S. Patent No. 6,465,136 (Fenton '136) is directed to a composite membrane structure comprising:

a composite membrane comprising a porous polymeric matrix and ionically conductive solid; and

at least one protective layer disposed adjacent to the to the composite membrane, comprising binder and ionically conductive solid.

It is apparent that the porous polymeric matrix (or binder) and ionically conductive solid in Fenton '136 correspond to components (B) (exactly the skeleton section of component (B)) and (C) in the present invention, respectively (although not specified by the Examiner in the Office Action).

However, Fenton '136 does not disclose a component corresponding to component (A), so that in Fenton '136, there is no description with respect to the covalent bond between the component corresponding to component (A) and the porous polymeric matrix (or binder).

Further, in Fenton '136, there is no description with respect to the compatibility between the porous polymeric matrix (or binder) and ionically conductive solid (more specifically, the requirement that $(\delta p^2 + \delta h^2)^{1/2}$ value for the porous polymeric matrix (or binder) is 7 or less). That is, in Fenton '136, the importance of this compatibility is not recognized at all.

As apparent from the above, Fenton '136 does not teach or suggest the essential features and

the unexpected effects of the present invention.

**IX. Claims 1-26 with claim 1 as the main invention are rejected under 35 U.S.C. 103(a) as being unpatentable over Chisholm et al. (U.S Patent no. 6,468,684).**

Claim 3 of U.S. Patent No. 6,468,684 (Chisholm et al.) is directed to a proton-conducting membrane comprising:

a solid acid material represented by a specific formula, and

a structural binder with said solid acid material wherein said structural binder is electrically conducting.

It is apparent that the structural binder (which may be a polymer) and solid acid material in Chisholm et al. correspond to components (B) (exactly the skeleton section of component (B)) and (C) in the present invention, respectively (although not specified by the Examiner in the Office Action).

However, Chisholm et al. does not disclose a component corresponding to component (A), so that in Chisholm et al., there is no description with respect to the covalent bond between the component corresponding to component (A) and the structural binder.

Further, in Chisholm et al., there is no description with respect to the compatibility between the structural binder and solid acid material (more specifically, the requirement that $(\delta p^2 + \delta h^2)^{1/2}$ value for the structural binder is 7 or less). That is, in Chisholm et al., the importance of this compatibility is not recognized at all.

As is apparent from the above, Chisholm et al. does not teach or suggest the essential features and the unexpected effects of the present invention.

**X. Claims 1-26 with claim 1 as the main invention are rejected under 35 U.S.C. 103(a) as being unpatentable over McGrath et al. (6,465,136) ([sic], should be US Patent Publication no. 2002/0091225).**

Claim 32 of U.S. Patent Application Publication No. 2002/0091225 A1 (McGrath et al.) is directed to a proton exchange membrane comprising a sulfonate copolymer represented by a specific formula. As is apparent from claim 33 of McGrath et al., this proton exchange membrane may further comprise an inorganic heteropoly acid.

It is apparent that the sulfonated copolymer and inorganic heteropoly acid in McGrath et al. correspond to components (B) (exactly the skeleton section of component (B)) and (C) in the present invention, respectively (although not specified by the Examiner the Office Action).

However, McGrath et al. does not disclose a component corresponding to component (A), so that in McGrath et al., there is no description with respect to the covalent bond between the component corresponding to component (A) and the sulfonated copolymer.

Further, in McGrath et al., there is no description with respect to the compatibility between the sulfonated copolymer and inorganic heteropoly acid (more specifically, the requirement that $(\delta p^2 + \delta h^2)^{1/2}$ value for the sulfonated copolymer is 7 or less). That is, in McGrath et al., the importance of this compatibility is not recognized at all.

As apparent from the above, McGrath et al. does not teach or suggest the essential features and the unexpected effects of the present invention.

**XI. Fenton et al. (6,638,659) has about the same teachings and suggestions as those in above applied Fenton et al. (6,465,136). Kim et al. US 2003/0099874 is cited to show prior art.**

Claim 1 of U.S. Patent No. 6,638,659 (Fenton '659) is directed to a process for fabricating a membrane electrode assembly (MEA) comprising:

a) obtaining a composite membrane having a first major surface area and a second major surface area comprising:

1) membrane layer containing ionically conductive solid and an ionomeric binder;

2) at least one protective layer disposed adjacent to the membrane layer comprising an ionomeric binder and an ionically conductive solid, and

optionally a hygroscopic fine powder;

b) applying a mixture of oxidizing catalyst, ionomeric binder and ionically conductive solid in a solvent on said first major surface area;

c) applying a mixture of reducing catalyst, ionomeric binder and ionically conductive solid in a solvent on said second major surface area.

Applicants note that Fenton '659 is a continuation-in-part of Fenton '136, cited in paragraph no. VIII. of the Office action.

It is apparent that the ionomeric binder and ionically conductive solid in Fenton '659 correspond to components (B) (exactly the skeleton section of component (B)) and (C) in the present invention, respectively (although not specified by the Examiner in the Office Action).

However, Fenton '659 does not disclose a component corresponding to component (A), so that in Fenton '659, there is no description with respect to the covalent bond between the component corresponding to component (A) and the ionomeric binder.

Further, in Fenton '659, there is no description with respect to the compatibility between the ionomeric binder and ionically conductive solid (more specifically, the requirement that $(\delta p^2 + \delta h^2)^{1/2}$ value for the ionomeric binder is 7 or less). That is, in Fenton '659, the importance of this compatibility is not recognized at all.

As apparent from the above, Fenton '659 does not teach or suggest the essential features and the unexpected effects of the present invention.

If, for any reason, it is felt that this application is not now in condition for allowance, the Examiner is requested to contact Applicants undersigned agent at the telephone number indicated below to arrange for an interview to expedite the disposition of this case.

In the event that this paper is not timely filed, Applicants respectfully petition for an appropriate extension of time. Please charge any fees for such an extension of time and any other fees which may be due with respect to this paper, to Deposit Account No. 01-2340.

Respectfully submitted,

ARMSTRONG, KRATZ, QUINTOS, HANSON & BROOKS, LLP

Daniel A. Geselowitz, Ph.D.
Agent for Applicants
Reg. No. 42,573

DAG/plb:mla

Atty. Docket No. **011786**
Suite 1000
1725 K Street, N.W.
Washington, D.C. 20006
(202) 659-2930

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